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Calculation of the viscosity of liquid Na-K and Na-Cs alloys

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Abstract A simple formalism relating thermodynamic and transport properties in liquid binary alloys has been applied to determine the viscosity of liquid alkali alloys. The formalism reproduced a qualitative trend of the estimated values of viscosity but there appears to be a significant quantitative disparity for this class of alloys

Keywords Liquid alloys, concentration fluctuation, viscosity

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A great deal of effort has gone into the understanding of the statics and dynamics of metallic liquid alloys. These studies include the investigation of the structural, thermodynamic and transport properties of the alloys. Many mearsurements have been made on the thermodynamic properties of liquid binary alloys [1] and some measurements on the transport properties of some metals and alloys [2]. However, very little investigation has been made on the relationship between thermodynamic properties and transport properties. Neale and Cusack [3] attempted an investigation with a view to obtaining a comprehensive empirical description between thermodynamic and transport properties in Na-Cs liquid alloys. A theoretical attempt to link thermodynamic properties of liquid binary alloys to some transport properties has been made by Singh and Sommer [4]. In their work, they presented a simple formalism which relates viscosity and diffusion coefficient to the thermodynamic properties of metallic alloy. This kind of formalism if it is in reasonable agreement with experimental measurements, could serve as a unique economic and effective method to predict the properties of liquid metallic binary alloys, depending less on experimental data.

Akinlade, Singh and Sommer [5] have worked on this formalism and applied it in calculating the viscosity of liquid Cu-Bi and Bi-Zn alloys by using energetics determined from thermodynamic data. The results appear impressive as it showed that for alloys of Bi with large size difference, the predictive power of the model is decreased towards some concentration range, and for alloys of Bi with size ratio close to one, the calculated viscosity values is reasonably close to the measured and estimated values of viscosity. In view of these results, it becomes of interest to apply the formalism on some other class of alloys. Here, this formalism is applied to alloys of simple metals (the alkali metals) which have phase seperating tendencies. The study will help to determine the extent of applicability of the formalism and the size ratio effects on the predictive power of the formalism for this class of alloys.

In particular, the viscosity of Na-K and Na-Cs liquid alloys are being predicted using energetics from thermodynamic investigations. A detailed description of the formalism used in this investigation is already given in Refs. [4,5]. The measured values of $S_{cc}(0)$ and free energy of mixing G_m/RT were reproduced by varying the free parameters in the expressions for $S_{cc}(0)$ and free energy of mixing as already given in Refs. [4,5] which are γ, the size ratio and W, a measure of the order energy. It is possible to obtain γ from experimental density measurements [5]. However in this work, it is choosen as a free parameter because experimental measurements for the densities at the working temperature are not available to the author.

Firstly, the values of γ and Ware chosen such that it reproduces the measured value of $S_{cc}(0)$ for the alloy. The experimental value of $S_{cc}(0)$ for Na-K alloy was obtained by making a polynomial fit of the activity data obtained from Ref. [6] and used in [7]. The derivative of the activity was obtained

and used in the expression for $S_{cc}(0)$ as already used in [8]. The experimental $S_{cc}(0)$ for Na-Cs was obtained directly from Ref. [3].

Figure 1 shows the calculated and measured values for the $S_{cc}(0)$. The figures show excellent agreement for Na-K and a good agreement for Na-Cs alloy. The largest disparity was observed between 0.6 and 0.8 atomic fraction of Na for the Na-Cs alloy. The values of γ and W that was used to reproduce the measured Scc(0) are $\gamma = 1.09$, $W = 0.91$ for Na-K and $\gamma = 2.76$, $W = 1.16$ for Na-Cs.

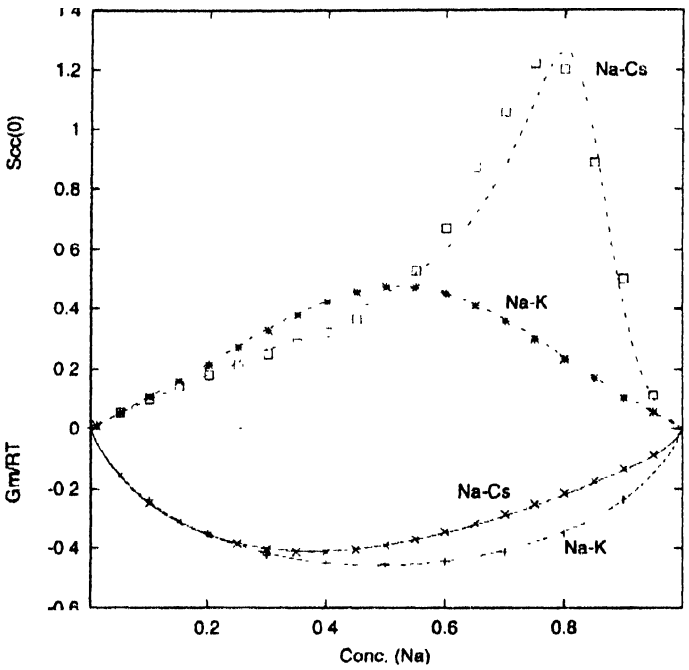

Figure 1. Comparism of experimental and computed values of $S_{cc}(0)$ and Gm/RT for Na-K and Na-Cs liquid alloys (. , ,) are experimental values of $S_{cc}(0)$ for Na-Cs estimated from Ref.[3]. (* * *) - are experimental $S_{cc}(0)$ for Na-K estimated from Ref.[6] (. - . - .) - are calculated values of $S_{cc}(0)$ for Na-Cs and Na-K alloys (X X X) are experimental values of Gm/RT for Na-Cs from Ref [3] (+ + +) are experimental values of Gm/RT for Na-K from Ref [6]. (. .) - calculated values of Gm/RT for Na-Cs. (- - -) - calculated values of Gm/RT for Na-K. (.....) are ideal values of $S_{cc}(0)$.

Using the above values of γ and W, the free energy of mixing is calculated and compared with the measured values. This is shown in Figure 1. The experimental values of the free energy of mixing is obtained from Ref. [6] for Na-K and Ref.[3] for Na-Cs. Here also, the figures show excellent fit for both Na-K and Na-Cs alloys. This points to the fact that this formalism can describe effectively the thermodynamic properties of sodium-based alkali alloys.

The viscosity coefficient $\Delta\eta / \eta_0$ was calculated for the two alloys using the parameters determined and the results are shown in Figure 2. The experimentally measured values of viscosity for these alloys were not available to the author; hence, the calculated values of viscosity which depend on γ and W, were compared with the viscosity values determined from experimental enthalpy of formation. This approach is expressed in Refs. [4,5]. The results of Akinlade *et al* [5] show that viscosity values determined from the experimental values of enthalpy of formation qualitatively agree and compare reasonably with measured viscosities. The enthalpy values used for Na-K was obtained from Ref.[6] and that used for Na-Cs was obtained from Ref.[9]. All measurements for both alloys were made at 384K.

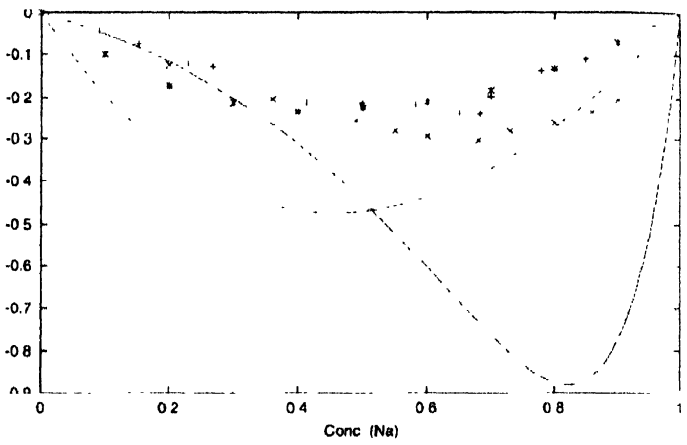

Figure 2. $\Delta\eta / \eta^0$ *vs* concentration of Na for Na-K and Na-Cs liquid alloys. (------) - calculated values for Na-Cs (+ + +) - estimated values from data of Ref [9] for Na Cs (X X X) - estimated values from data of Ref. [10] for Na-Cs (- - -) - calculated values for Na-K liquid alloy (* * *) - estimated values from data of Ref [6] for Na-K

The results in Figure 2 show that for Na-K which has a size ratio close to one, the calculated values of viscosity show qualitative trend with the viscosity values determined from measured enthalpy data. However, the disparity is wide when compared to the result of Akinlade *et al* [5] for Cu-Bi which also has a size ratio close to one hence suggesting a limitation to the applicability of the formalism for alkali-based alloys. The calculated values for Na-Cs show some level of closeness with the viscosity values determined from the measured enthalpy values at low concentration of Na. At high concentration of Na, a large disparity is observed, but this disparity is consistent with that observed for Cu-Bi alloy [5] with a large size ratio. This large disparity and skewed form of the calculated values may be a result of the size factor since the difference between the value of W for Na-K and the value of W for Na-Cs is not large.

It can therefore be mentioned that this simple formalism proposed by Singh and Sommer [4] has been used with some level of success to determine the viscosity of some liquid binary alloys. Its application to Na-K and Na-Cs gave a qualitative trend to the measured viscosity with a significant quantitative disparity.

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